

Argonne National Laboratory

**THE CONTINUOUS MONITORING OF
BORIC ACID CONCENTRATION
BY NEUTRON ABSORPTIOMETRY**

by

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Reactor Engineering Division

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ABSTRACT

A neutron absorption cell has been built to monitor continuously the boric acid concentration of reactor water flowing through a purification loop. The instrument has been calibrated over the range from 0 to 2.65 gm/liter with a maximum absolute error of 0.05 gm/liter.

I. INTRODUCTION

In the operation of the EBWR at higher power levels (20 to 100 Mwt), boric acid will be used to provide a reactivity margin for shutdown.⁽¹⁾ Several means are available for monitoring boric acid concentration in the moderator, including chemical titration, photometric determination, and a neutron absorption technique.

The neutron absorption technique is feasible for the determination of soluble poisons in water and a number of other solvents. This method was investigated for the monitoring of boric acid concentration in the EBWR water moderator, and an instrument was developed on the principle of neutron absorptiometry which meets the following conditions:

- (1) The instrument operates over a concentration range from 0 to 2.65 gm of boric acid per liter of water.
- (2) The sensitivity of the instrument is 0.05 gm of boric acid per liter of water.
- (3) It is capable of monitoring the boric acid concentration in a flowing sampling stream of reactor water.
- (4) The instrument operates comparatively independent of temperature.
- (5) It is stable and results are reproducible.
- (6) The operation of the instrument is simple.

II. DEVELOPMENT AND DESIGN

Work previously done with the technique involved the use of BF_3 counting chambers with a suitable neutron source.^(2,3) The geometry was such that the source neutrons were moderated and passed through the liquid sample before reaching the detector. The samples were then counted, and the poison concentration was related to the counting rate. In the development of the completely continuous monitoring system, the counting chamber was replaced by a BF_3 ionization chamber.

Preliminary measurements were made by placing a BF_3 ionization chamber in the center of a tank of water and a source of neutrons on the outside edge of the tank. A small current was observed, and the addition of boric acid to the water caused enough reduction in the current to continue investigation on this method.

Geometries were investigated to optimize current in the BF_3 chamber and sensitivity to the introduction of boric acid. A circular tank, 30.5 cm (12 in.) in diameter and 45.8 cm (18 in.) high, was fabricated so that the detector could be fixed in the center, and the source could be moved inside the tank closer to or away from the detector. The tank was filled with water, and known amounts of boric acid were added. The change in electrometer output as a function of source position was noted (Figure 1 and Table I).

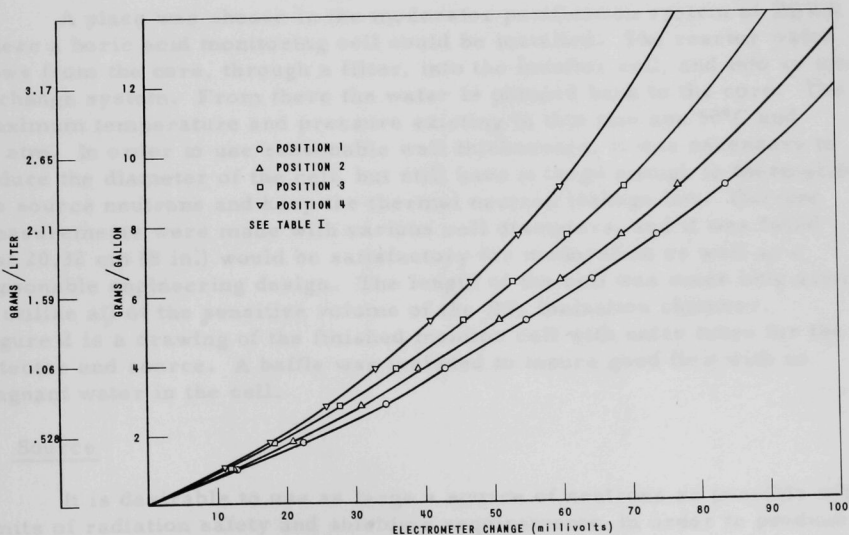


FIG. 1
BORIC ACID CONCENTRATION VS. CHANGE IN ELECTROMETER
OUTPUT PARAMETRIC WITH SOURCE POSITION

This information shows that for those distances where measurements were made, the fractional current change is independent of the distance from source to detector. The maximum current is obtained when the source and detector are as close together as possible.

Table I

CURRENT AS A FUNCTION OF SOURCE TO DETECTOR DISTANCE

Position	Source to Detector Distance, Center to Center		I_0^* (amp)
	(cm)	(in.)	
1	5.08	2	0.4×10^{-9}
2	7.62	3	0.37×10^{-9}
3	10.16	4	0.33×10^{-9}
4	12.70	5	0.28×10^{-9}

Note: See Figure 1.

*Current with 0 concentration

A place was chosen in the moderator purification system of EBWR where a boric acid monitoring cell could be installed. The reactor water flows from the core, through a filter, into the monitor cell, and into an ion exchange system. From there the water is pumped back to the core. The maximum temperature and pressure existing in this line are 50°C and 42 atm. In order to use reasonable wall thicknesses, it was necessary to reduce the diameter of the cell, but still have it large enough to thermalize the source neutrons and keep the thermal neutron leakage low. Current measurements were made with various cell diameters, and it was found that 20.32 cm (8 in.) would be satisfactory for moderation as well as a reasonable engineering design. The length of the cell was made long enough to utilize all of the sensitive volume of the BF_3 ionization chamber. Figure 2 is a drawing of the finished monitor cell with entry tubes for the detector and source. A baffle was included to insure good flow with no stagnant water in the cell.

A. Source

It is desirable to use as large a source of neutrons as possible within limits of radiation safety and shielding requirements, in order to produce a large current and minimize statistical fluctuations. A long half-life would reduce the change in current with time and the need for constant calibration of the instrument.

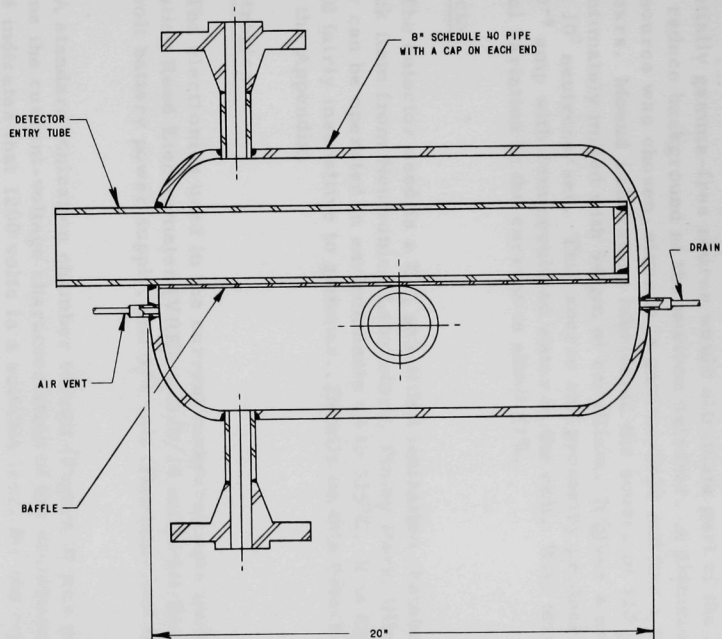
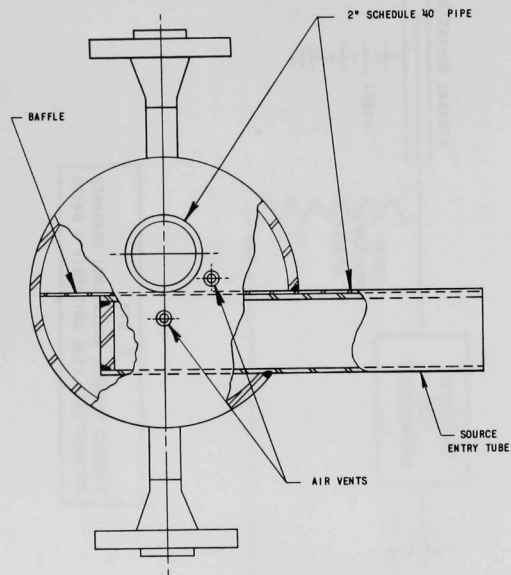


FIG. 2
BORIC ACID MONITOR CELL

An essentially gamma-free source would eliminate part of the health hazard and also reduce background at the neutron detector. A plutonium-beryllium neutron source was chosen, since it is gamma-free and has a half life of 24,600 years. Mound Laboratory fabricated the source of 110 gm of plutonium intimately mixed with 54 gm of beryllium. It gives a neutron output of 1.28×10^7 neutrons/sec. This source and geometry produce a current of 0.66×10^{-9} amp with demineralized water in the cell. With this current, the statistical variation in the current is about $\pm 1\%$.

B. Detector

The detector used is a BF_3 gas-filled ionization chamber purchased as a stock item from Neutronics Laboratory, Tinley Park, Illinois. This chamber can be operated in environments up to 315°C . It is highly shock-proof and fairly insensitive to gammas. Details on this detector may be found in the Appendix.

C. Electronics

The electronics used in the current measurements includes a Model 30 Vibrating Reed Electrometer (VRE), a 1/0/10 millivolt Brown Recorder, a 1200-volt battery power supply, and 0/1 volt transistorized feedback supply.

A standard ionization chamber circuit (Figure 3) was used to determine the current-voltage characteristics of the ionization chamber. Figure 4 indicates that 1200 volts is a suitable level for the high-voltage supply.

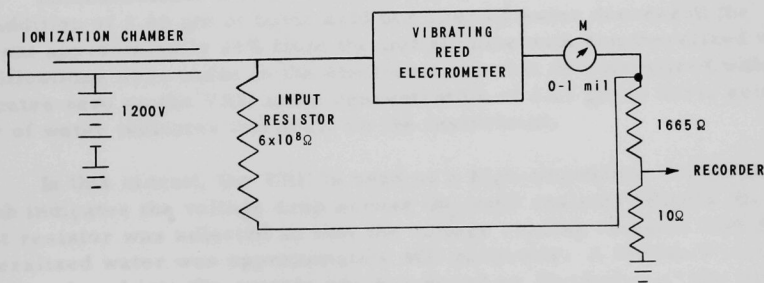


FIG. 3
STANDARD IONIZATION CHAMBER CIRCUIT
USING A VIBRATING REED ELECTROMETER

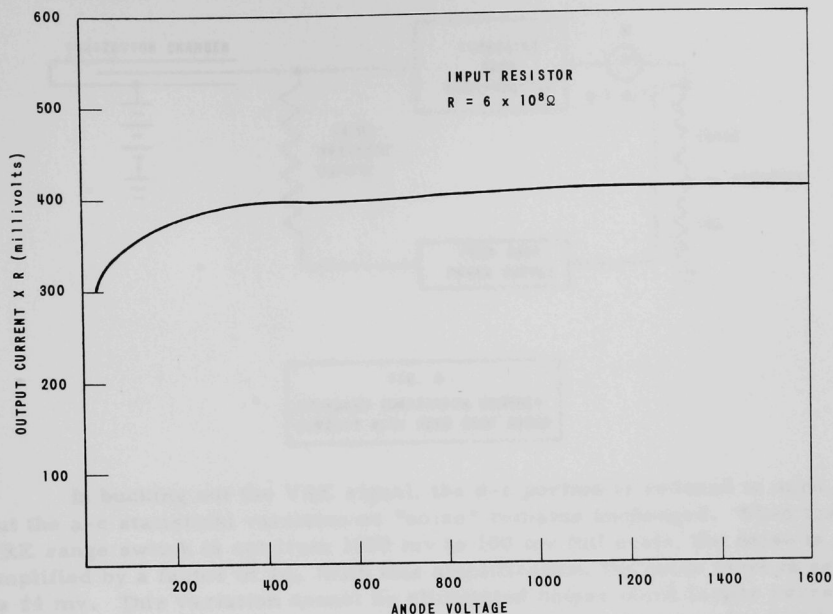


FIG. 4
CURRENT-VOLTAGE CHARACTERISTICS
OF THE NEUTRON IONIZATION CHAMBER

Measurements with and without boric acid in the cell indicated that the addition of 2.65 gm of boric acid per liter of water decreased the current approximately 25% from the initial value with demineralized water. Modifications were made in the electronics so that demineralized water indicates zero on the VRE and a concentration of 2.65 gm of boric acid per liter of water indicates full scale on the instrument.

In this circuit, the VRE is used as a high-impedance voltmeter which indicates the voltage drop across the input resistor (Figure 3). The input resistor was selected so that the voltage reading obtained with demineralized water was approximately 400 millivolts. A feedback circuit was introduced into the system which reduced or "bucked-out" the voltage on the electrometer to zero. A concentration of 2.65 gm of boric acid per liter of water results in a meter reading of -100 millivolts. A meter reversal switch on the electrometer reverses the polarity, and the meter reads upscale rather than below zero. The instrument reads zero for demineralized water and 100 millivolts for a concentration of 2.65 gm per liter. The range switch on the VRE is set on 100 millivolts full scale, so that a full-scale deflection is observed at maximum concentration. Figure 5 shows the circuit for the modified system.

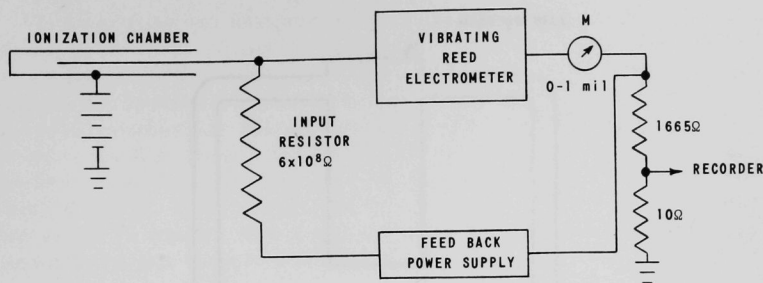


FIG. 5
STANDARD IONIZATION CHAMBER
CIRCUIT WITH FEED BACK ADDED

In bucking out the VRE signal, the d-c portion is reduced to zero, but the a-c statistical variation or "noise" remains unchanged. When the VRE range switch is set from 1000 mv to 100 mv full scale, the noise is amplified by a factor of 10. With this amplification, the noise level is seen as ± 4 mv. This variation cannot be eliminated unless much larger currents are produced. Since it is not feasible in this system, the recorder has been mechanically damped to allow the signal displayed on the chart to be more easily read. A 50-rpm motor was substituted for the normal pen balance motor (180 rpm). This reduced the high-frequency response of the recorder and reduced the noise to ± 2 mv.

Because of the noise, it is difficult to zero the VRE with the panel meter. The recorder is used, and the signal trace gives an accurate zero adjustment. The -1/0/10 range is used since this moves the recorder zero one unit from the edge of the chart, so that the zero trace may clearly be seen.

The recorder was checked with a potentiometer and found to be linear. A check of the panel meter on the VRE showed it was not linear. For this reason all measurements were made by reading the recorder and ignoring the panel meter on the VRE.

III. STABILITY AND TEMPERATURE MEASUREMENTS

A loop was set up so that water or a solution of boric acid could be pumped through the boric acid monitor cell (Figure 6). Stability, temperature, and calibration measurements were made with this system.

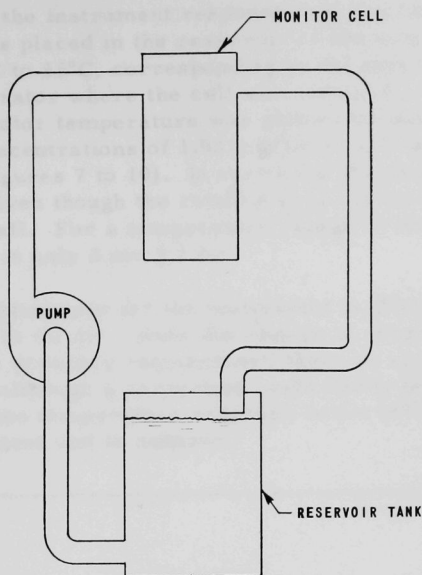


FIG. 6
LOOP SETUP USE FOR TESTING AND
CALIBRATING THE BORIC ACID MONITOR

The stability of the system depends upon the stability of the ionization chamber power supply, the VRE, and the feedback voltage supply. Experience with the electrometer has shown it to be very stable. The ionization chamber is being operated well on its plateau, so that small voltage variations cause no problem. The feedback voltage must be very stable because a 1% change in the feedback voltage can cause a net change of as much as 10 mv or an error of ± 0.26 gm per liter. A transistorized voltage supply modified to have a variable output of 0 to 1 volt is used for this circuit. Previous measurements indicated that the output should vary not more than ± 0.1 mv in 1 volt.

The stability check of the entire system was made in the following manner: The circuitry was assembled, and demineralized water was pumped through the system. A zero reading on the recorder was obtained with the feedback power supply, and it was left in this condition for three days. At the end of this time, the recorder trace was inspected, and the maximum drift that was observed was less than 1 mv. This test was repeated several times with the same result, indicating that the system is quite stable.

To determine the instrument response to temperature variation, an immersion heater was placed in the reservoir of the loop. The temperature was varied from 25°C to 55°C, corresponding to the expected temperature variation of the moderator where the cell was installed. Instrument response versus moderator temperature was plotted for demineralized water, and for boric acid concentrations of 1.02 gm/liter, 1.43 gm/liter, and 2.51 gm/liter (see Figures 7 to 10). In each case the slope of the line was $-0.05 \text{ mv}/^\circ\text{C} \pm 50\%$. Even though the relative error in this is large, the absolute error is small. For a temperature change of 40°C, the change in instrument response is only $2 \text{ mv} \pm 1 \text{ mv}$.

The error requirement for the instrument is $\pm 0.05 \text{ gm/liter}$, which corresponds roughly to $< 2 \text{ mv}$. Since the change in response with temperature is less than the accuracy requirement, there is no need for temperature compensation, although a correction could easily be made if desired. It is noted here that the temperature response below 25°C and above 55°C has not been investigated and is unknown.

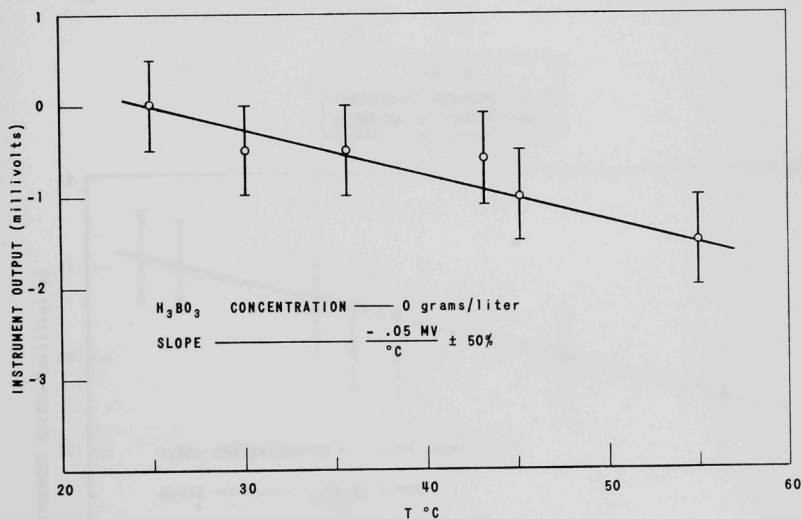


FIG. 7
INSTRUMENT RESPONSE AS A
FUNCTION OF TEMPERATURE

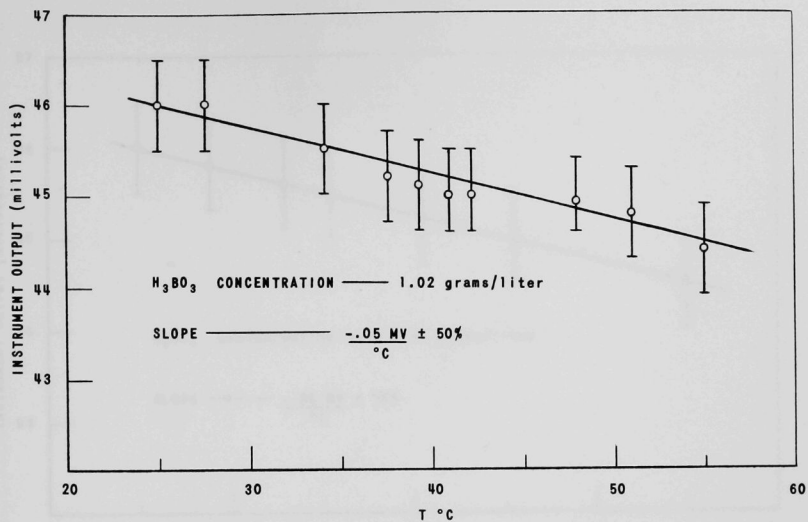


FIG. 8
INSTRUMENT RESPONSE AS A
FUNCTION OF TEMPERATURE

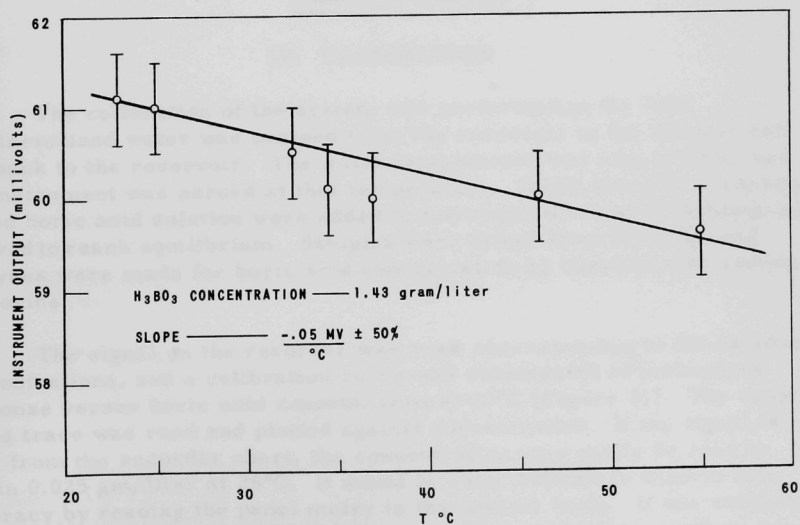


FIG. 9
INSTRUMENT RESPONSE AS A
FUNCTION OF TEMPERATURE

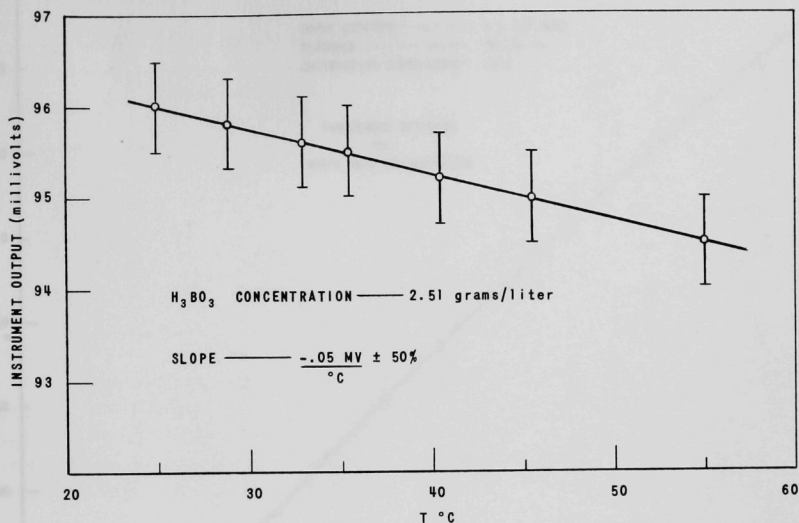


FIG. 10
INSTRUMENT RESPONSE AS A
FUNCTION OF TEMPERATURE

IV. CALIBRATION

The calibration of the system was performed in the loop. Demineralized water was pumped from the reservoir to the monitor cell and back to the reservoir. The water temperature was kept at 25°C, and the instrument was zeroed at this temperature. Small amounts of concentrated boric acid solution were added to the reservoir, and the system was allowed to reach equilibrium. Samples were drawn from the loop, and analyses were made for boric acid concentration by titration with sodium hydroxide.

The signal on the recorder was read corresponding to the various concentrations, and a calibration curve was constructed of instrument response versus boric acid concentration at 25°C (Figure 11). The center of the trace was read and plotted against concentration. If the signal is read from the recorder chart, the concentration may easily be read to within 0.025 gm/liter at 25°C. It would be very difficult to achieve this accuracy by reading the panel meter in the control room. If one watches the needle variations for a few moments and averages them, perhaps an accuracy of ± 0.05 gm/liter may be read. To read with an accuracy of ± 0.025 gm/liter, the recorder chart must be read. If the temperature is above 25°C and below 55°C, the concentration may be corrected to 25°C by using the temperature coefficient of -0.05 mv/°C indicated above.

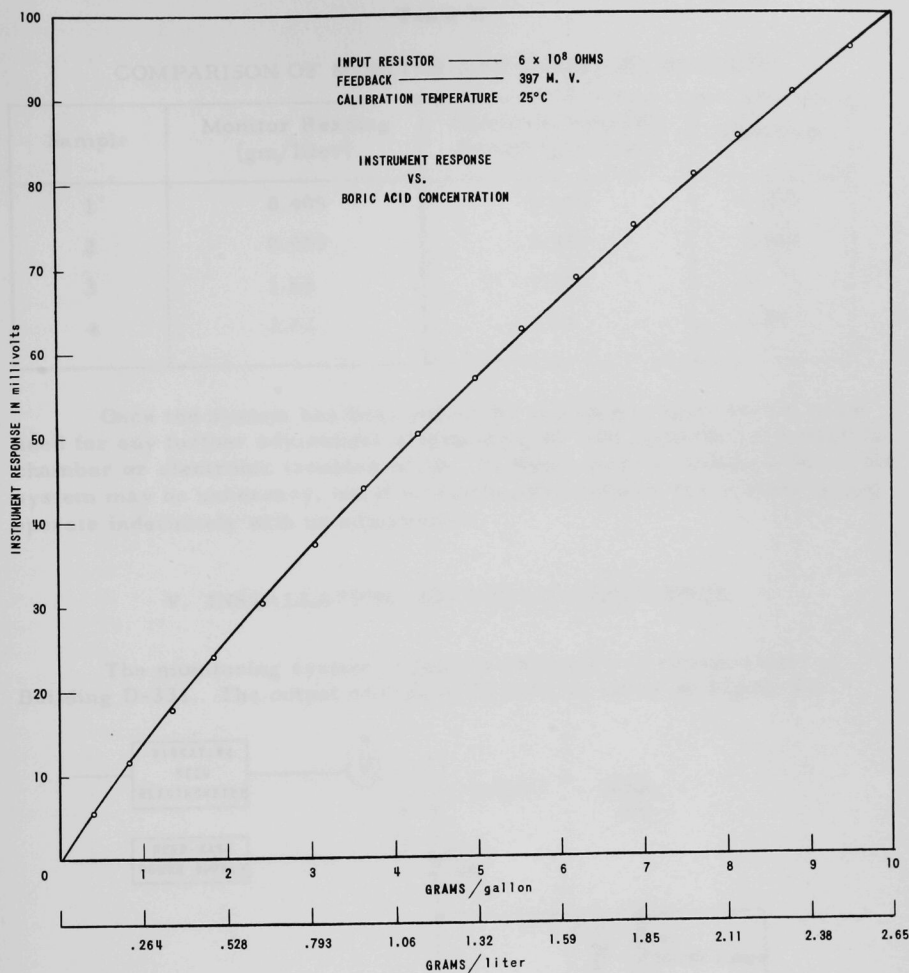


FIG. II
 MONITOR CALIBRATION CURVE

The monitor cell and associated electronics were set up in the test loop and calibrated with known concentration of boric acid. The system was checked periodically by additions and removal of boric acid from the loop. The loop concentration was tested chemically and compared with the results obtained from the monitoring system. The agreement between the two was very good, as shown by samples in Table II.

Table II

COMPARISON OF MONITOR AND CHEMICAL RESULTS

Sample	Monitor Reading (gm/liter)	Chemical Analysis Result (gm/liter)	Deviation
1	0.409	0.394	0.015
2	0.970	0.983	0.013
3	1.64	1.64	0
4	2.52	2.51	0.01

Once the system has been set up for operation, there should be no need for any further adjustment unless changes take place in the ionization chamber or electronic troubles occur. In those cases a recalibration of the system may be necessary, but if no malfunction occurs, the system should operate indefinitely with no adjustments.

V. INSTALLATION AND INITIAL EXPERIENCE

The monitoring system is located physically in the basement of Building D-331. The output circuit of the VRE is shown in Figure 12.

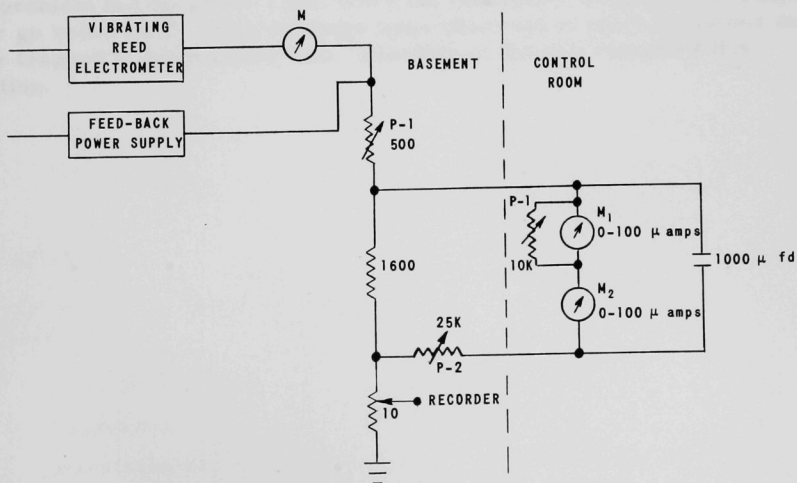


FIG. 12
MODIFIED OUTPUT CIRCUIT OF
VIBRATING REED ELECTROMETER

Meters m_1 and m_2 are 100-microampere movements and are adjusted by means of p_1 , p_2 , and p_3 to read the same as the recorder and meter m . A meter relay m_1 to which the alarm system is connected is located in the control room, and m_2 is a panel meter (also located in the control room) which displays the concentration signal to the reactor operators. A 100- μ f capacitor was connected across meters m_1 and m_2 to damp the rather wild needle movements which occur due to the statistical variation in the signal.

A number of situations may arise which will cause the monitor to give erroneous readings. The presence of neutron-absorbing material other than boric acid in the moderator will cause the monitor to read high. Extraneous neutron sources or an unusually high neutron or gamma background will cause the monitor to read low. Air trapped in the monitor cell would give a high reading since moderator is displaced by air. This can be overcome by bleeding the cell periodically. Temperatures below 25°C or above 55°C may also give trouble, since no calibration measurements were made beyond these limits.

Since the installation of the boric acid monitoring system in the EBWR, it has been placed in operation during hot and cold critical runs where boric acid was used in the reactor water. Instrument readings were compared with chemical tests on samples drawn from the monitor cell, and the system was seen to be functioning properly. During cold critical runs the moderator temperature in the monitor cell was as low as 12°C, which resulted in high instrument readings. A temperature correction reduced the reading to within the required instrument error. This is not expected to be a problem during power runs, since the moderator temperature should never go below 25°C. High readings were observed at other occasions due to air trapped in the monitor cell. Bleeding of the cell remedied this situation.

APPENDIX

Neutron Source

Source No.	M-856
Neutron Output	1.28×10^7 neutrons/sec
Plutonium Content	110.02 gm
Beryllium Content	54.01 gm
Half-life	24,600 yr
Outer Jacket Material	Type 304 stainless steel
Outer Dimensions	3.94 cm x 6.57 cm long
Temperature	May be maintained at 500°C indefinitely
Manufacturer	Mound Laboratory Miamisburg, Ohio

DetectorNeutron Ion Current Chamber -
Model NL-IC2G

Dimensions	4.13 cm diam x 55.6 cm long
Sensitivity to Neutrons	2.15×10^{-13} amp/nv
Sensitivity to Gammas	1.9×10^{-11} amp/r/hr
Signal to Case Leakage at 315°C	10^{12} ohms
Signal to Case Capacity	30 micromicrofarads
Manufacturer	Neutronics Laboratory Tinley Park, Illinois

ElectrometerVibrating Reed Electrometer -
Model 30

Input Impedance	10^{16} ohms
Sensitivity	10^{-16} amp
Manufacturer	Applied Physics Corporation Pasadena, California

Feedback Power Supply

Transistorized d-c Power Supply -
Model P98-58462

Output

0/1 volt variable
9.0 volts fixed

Manufacturer

Electronics Division
Argonne National Laboratory

Recorder

Brown Strip Chart Recorder

Range

-1/0/10 millivolts

Manufacturer

Minneapolis Honeywell
Philadelphia, Pennsylvania

Monitor Cell

Fabricator
(see Figure 2)

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